

# PATENT SPECIFICATION

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## COMPLETE SPECIFICATION

### Improvements relating to Pigment Masses

I, SAMUEL NASE HUNTER, a citizen of the United States of America, of 639, Veronica Street, East St. Louis, State of Illinois, United States of America, do hereby declare the invention, for which I pray that a patent may be granted to me, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to paste pigments wherein finely divided metal or non-metal pigment matter, are wetted and mixed with certain organic compounds to form paste-like masses, hereinafter called paste pigments. The paste pigments of this invention are all characterized by being non-hygroscopic, non-corrosive, non-hydrolyzing in the presence of water, mineral acids or alkalis, and non-discoloring, and by remaining usable indefinitely without special care in storage, and by the preservation and enhancement of the brilliance of the pigment particles.

While this invention is not confined thereto, it has particular application to metallic pigments. Metallic paste-like pigments, especially, aluminium paste pigments, are known to the art. However, the metallic paste pigments known heretofore have had several disadvantages. The wetting agents used in paste pigments known heretofore have been highly volatile, hygroscopic and inflammable. Special care has had to be used in the handling and storage of such paste pigments. Caking, hardening and hard packing of the pigments with exposure to the atmosphere have been major problems. At the same time, the presence of moisture in such paste pigments caused internal pressure in the sealed containers in which they must be stored. The tendency for metallic pigments to oxidize, tarnish and discolor with the wetting agents of the paste pigments known heretofore, has, as a practical matter, restricted the use of metallic pigments to aluminium, though even aluminium has

not developed its full brightness in such paste pigments.

The paste pigments known heretofore have been entirely incompatible with lacquers, with certain resinous vehicles (such, for example, as polyvinyl chloride), and with certain ink vehicles, among others, so that the fields of their use have been limited.

The paste pigments of this invention have no counterpart in commerce.

One of the objects of this invention is to provide a plastic pigment which, besides exhibiting all of the general characteristics recited above, is hydrophobic, remains soft and permanently useful indefinitely, and is highly heat-resistant.

In accordance with this invention, generally stated, a paste pigment is provided which comprises one or more metallic or nonmetallic (organic or inorganic) pigments (which may be in the form of finely divided particles, powder, dust flakes, or in other finely divided form) which have been wetted and coated with a wetting agent consisting of, or containing at least 5% (by weight) chlorinated polyphenyl in liquid form or in solution, the proportion of the said wetting agent used in the wetting of the pigment being within the range 15—90% (by weight) of the paste pigment formed therewith.

Among the metallic pigments which may be used are aluminium (either polished or unpolished), silver, gold, bronze (of any composition or alloy, either bright or russett), lead, antimony, nickel, titanium, copper, carbonyl iron, elemental iron, stainless steel, beryllium, zinc, tin, chromium, vanadium and tungsten. Various other metallic pigments may be used as well as mixtures of such pigments. The paste pigments of this invention have a higher degree of brightness, specular gloss and reflectivity than the paste pigments known heretofore, and the metallic pigments of this invention lose none of their lustre even on indefinitely prolonged exposure to the air and moisture

and corrosive environments. Paste pigments of copper and bronze, for example, have heretofore been entirely impractical because of their discoloration or oxidation. However, in the form of pigment masses of the present invention, copper, bronze, carbonyl iron (which has been unknown in paste pigments form heretofore), remain undisclored and plastic indefinitely, though exposed to moisture and to corrosive environments such as acid and alkaline atmospheres. Such materials as bentonite, slate dust, silica, steatite and pyrophyllite may be used, and are useful in many application such as paints, roofing compounds and brick and concrete coatings. Black or red copper sulphide is used for main applications. Barium sulphide is used as an ingredient in producing luminous paints. Carbon blacks are used to produce a glossy black paste pigment, as does levigated charcoal. Barium and lead carbonates are among other various pigment particles which are used. Other lead pigments, particularly useful in acid environments are the antimonate, bichromate, chromate, and iodide. Finely divided graphite, natural or synthetic, is used to form paste pigments with numerous uses, such as thread dressings, and paints. Ultramarine produces a bright blue pigment mass with the wetting agents of this invention.

Such materials as cuprous oxides and copper pentachlorophenate may be incorporated in the paste pigments to meet

requirements of special applications such as marine paints, coatings and the like.

The wetting agents employed in the paste pigments of this invention contain chlorinated compounds (or combinations of chlorinated compounds) which have the general character of chlorinated biphenyl (diphenyl). Chlorinated biphenyls of different and desirable chlorine contents, or various other chlorinated polyphenyls (with or without solvents, polar; non-polar or chlorinated) may be employed.

The chlorinated biphenyls (diphenyls) and the chlorinated polyphenyls containing at least two linked phenyl radicals (such as chlorinated terphenyl (chlorinated 1,2-, 1,3- and/or 1,4-diphenyl benzene), and all of the chlorinated tetraphenyls) are included within the generic term "chlorinated polyphenyls".

As a constituent of the wetting agents of this invention, one or more extenders, thinners, additives, diluents, solvents, or plasticizers may be employed, of which the following are examples: chlorinated oils, silicone resins; silicone solutions; alkylenes (unsaturated hydro-carbons); polyalkylene glycols and their derivatives; unsaturated petroleum polymers; furfural; diphenyl ether; furan; oleo resins; oleic acid; palmitic acid and and plasticizers such as dibutyl phthalate, amyl acetate and tri-cresyl phosphate.

Illustrative examples of suitable wetting agents with or without suitable polar or non-polar solvent are:—

- |     |      |          |  |
|-----|------|----------|--|
| 75  | (1)  | 75% 54%  | chlorine content chlorinated biphenyl  |
|     |      | 25% 48%  | " " " "  |
|     | (2)  | 60% 54%  | " " " "  |
|     |      | 40% 42%  | " " " "  |
|     | (3)  | 50% 54%  | " " " "  |
|     |      | 50% 32%  | " " " "  |
| 80  | (4)  | 90% 21%  | " " " "  |
|     |      | 10% 48%  | " " " "  |
|     | (5)  | 95%—100% | 21% chlorine content chlorinated biphenyl  |
|     |      | 5%—0%    | trichloroethylene  |
| 85  | (6)  | 95%—100% | 48% chlorine content chlorinated biphenyl  |
|     |      | 5%—0%    | trichloroethylene  |
|     | (7)  | 95%—100% | 54% chlorine content chlorinated biphenyl  |
|     |      | 5%—0%    | trichloroethylene  |
|     | (8)  | 70% 21%  | chlorine content chlorinated biphenyl  |
|     |      | 20% 48%  | " " " "  |
| 90  |      | 10% 54%  | " " " "  |
|     | (9)  | 25% 60%  | chlorine content chlorinated biphenyl (a light yellow soft, sticky resin) dissolved in |
|     |      | 75%      | trichloroethylene  |
| 95  | (10) | 20% 70%  | chlorine content chlorinated biphenyl dissolved in                                     |
|     |      | 80%      | trichloroethylene  |
|     | (11) | 75% 21%  | chlorine content chlorinated biphenyl (a colorless mobile oil)                         |
|     |      | 25%      | trichloroethylene  |
|     | (12) | 5% 60%   | chlorine content chlorinated terphenyl (a yellow transparent resin) dissolved in       |
| 100 |      | 95%      | trichloroethylene  |

60	(13)	30%	60%	chlorine content chlorinated terphenyl solvent, polar or non-polar
		70%		
	(14)	60%	21%	chlorine content chlorinated biphenyl
		20%		silicon oil
5		8%		non-polar solvent
25		12%		chlorinated paraffin oil
	(15)	50%	42%	chlorine content chlorinated biphenyl
		40%		Furan
		10%		Aromatic solvent
10	(16)	15%	15%	chlorine content chlorinated diphenyl ether
67		85%	48%	chlorine content chlorinated diphenyl
	(17)	10%	12%	oleic acid
		90%	88%	chlorine content chlorinated diphenyl
	(18)	15%		furan
15		15%	60%	chlorine content chlorinated terphenyl
		70%		trichloro ethylene
	(19)	5%	65%	chlorine content mixed chlorinated polyphenyl isomers
		20%	60%	chlorine content chlorinated terphenyl
		5%	42%	chlorine content chlorinated diphenyl
20		70%		xylene
7	(20)	25%	60%	chlorine content chlorinated terphenyl
		1.7%		tricresyl phosphate
		73.3%		diacetone solvent

25 All percentage values quoted in this specification for the amounts of the constituents of the wetting agents and for the chlorine content of the chlorinated polyphenyls will be understood to refer to percentages by weight.

30 In producing many of the paste pigments of this invention, it is not necessary to grind the pigment matter and wetting agent together. A simple cold or hot mixing process will suffice in most instances.

35 Prewetting of soft, porous, vesicular or colloidal and gelatinous pigment matter having high absorption coefficients is often desirable and advantageous. It assures thorough wetting of the pigment, and permits pregelling of gelatinous pigment matter.

40 Illustrative examples of paste pigments in accordance with this invention are as follows:—

#### EXAMPLE I.

A paste pigment useful as an ingredient of a large variety of products including paints, enamels, lacquers, inks and adhesives is made of the following components, by weight:—

50 54% chlorine content chlorinated biphenyl, 60 parts finely powdered aluminium, 40 parts.

55 The components are mixed to wet and coat the particles of pigment, to form a homogeneous composition by any suitable mixing means, as, for example, a muller or a tumbler. The paste pigment of this composition is stable at high temperatures, non-inflammable, acid-, alkali-, salt-resistant, brilliant, quick and thorough-leaving, and easily dispersed. It remains homogeneous, soft and usable

indefinitely though exposed to the 65 atmosphere and corrosive environments.

Paste pigments useful as ingredients of printing inks are illustrated by the compositions of Examples II—V as follows:—

#### EXAMPLE II.

Wetting agent 5 above, 85—90 parts  
Atomized Al Pigment, 15—10 parts  
Polar or chlorinated solvents may shorten the drying time.

#### EXAMPLE III.

Wetting agent 6 above, 85—90 parts  
Atomized Al Pigment, 15—10 parts.

#### EXAMPLE IV.

Wetting agent 7 above, 85—90 parts 80  
Atomized Al Pigment, 15—10 parts.

#### EXAMPLE V.

Wetting agent 8 above, 85—90 parts  
Carbon Black Pigment, 15—10 parts.  
Example V above is illustrative of the 85 use of mixtures of chlorinated biphenyls of different chlorine content. The proportions of the different components of the wetting agent may be varied to form useful pigments in fast drying putty, 90 caulking, or dressings, requiring low pigment concentrations.

#### EXAMPLE VI.

A paste pigment suitable for use in fast-drying paints or mastics without the 95 use of driers is made of the following components by weight:—

Wetting agent 12 above, 45—50 parts  
Bronzing powders Pigments, 55—50 parts. 100

Other paste pigments suitable for use in fast-drying paints or mastics without the use of driers may be made with wetting agents 9 and 10, 45—50 parts, bronzing  
5 Pigment, 55—50 parts.

#### EXAMPLE VII.

A paste pigment suitable for use in fast-drying interior paint without the use of driers may be made of the following components, by weight:—  
10 Wetting agent 11 above, 46—60%  
Colloidal clay Pigment, 54—40%.

#### EXAMPLE VIII.

A paste pigment suitable for use with  
15 polyvinyl chloride to form a highly reflecting pigmented film is made of the following components by weight:—  
Wetting agent consisting of 50% 48%  
20 50% 54% chlorine content chlorinated biphenyl, 46 parts  
Pigment (aluminium powder), 54 parts.  
The addition of 12% of the above paste pigment to the polyvinyl chloride is sufficient for some applications. Some will  
25 require polar solvent.

#### EXAMPLE IX.

A paste pigment suitable for use as a pipe thread compound or as an ingredient  
30 of paint is made of the following components by weight:—  
Wetting agent 1 above, 50 parts  
Graphite, 50 parts.

#### EXAMPLE X.

A paste pigment suitable for use as an ingredient of paint is made of the following components by weight:—  
35 Wetting agent 2 above, 46—60 parts  
Titanium dioxide, 54—40 parts.

#### EXAMPLE XI.

A paste pigment suitable for use in an ingredient of paints or as a coating composition is made of the following components by weight:—  
40 Wetting agent 3 above, 40—60 parts  
Elemental Iron, 54—50 parts.  
A convenient form of iron powder for use with this and similar formulations is the so-called Carbonyl Iron Powder.

#### EXAMPLE XII.

A paste pigment suitable for making luminous paint is made of the following components by weight:—  
50 54% chlorine content chlorinated biphenyl, 46—60 parts  
Pigment (Barium sulphide), 54—40 parts.  
Unlike the linseed oil-turpentine pastes of the prior art, this composition is stable,

non-oxidizing, non-inflammable and non-volatile.

#### EXAMPLE XIII.

A paste pigment suitable for use as a roofing compound is made of the following components by weight:—  
65 Wetting agent 1, above 40 parts  
Talc, steatite, or slate dust, 60 parts.  
Such a roofing composition has numerous advantages over asphaltic roofing compositions. It does not soften with heat, nor  
70 become brittle with age, and it is non-inflammable.

#### EXAMPLE XIV.

A paste pigment suitable for use in primer coatings is made of the following components by weight:—  
75 Wetting agent (14), 40—60 parts  
Copper powder pigment, 60—40 parts.  
Wetting agents (15), (16) and (17) may be substituted for wetting agent (14) to produce paste pigments having various characteristics consistent with the general characteristics of the pigment masses of this invention.

The examples given are, of course,  
85 merely illustrative. In paste pigments for use in paints, concentrations of pigment of 74% or more may be used, for example. The proportions of chlorinated polyphenyls of different chlorine content  
90 in the wetting agent, the proportions of wetting agent to pigment, and the character of the pigments may be varied. The substitution of other pigment described for the aluminium of Example 1, for  
95 instance, will yield a useful paste pigment. Mineral acid-resistant paste pigments which also give acid-resistant coatings, may be made up with mixtures of 21% and 48% chlorine content chlorinated biphenyl. Straight 48% chlorine  
100 content chlorinated biphenyl may be used to make up a salt-resistant paste pigment. The biphenyls of less than 54% are not entirely non-inflammable but burn with  
105 difficulty and will not support combustion when removed from an open flame.

In producing the putty-like paste pigments of this invention, a wetting agent of the character here disclosed is taken  
110 up in an organophilic cation-modified onium bentonite or the character described in British Patent No. 664,830. The wetting agent may be further pigmented with metal, non-metal or gelatinous pigment matter before being taken  
115 up or absorbed by the gel-forming pigment matter. In any event, the organophilic modified clay pigment swells and takes up the wetting agent to form a smooth, uniform composition  
120 which does not subsequently lump or oxidise, but remains plastic and permanently useful.

Various fillers, extenders and additives, such as calcium carbonate, slate dust, stearite, pyrophyllite, carbon black and amorphous silica may be added to the paste pigment compositions to reduce production costs, obtain various colors and other preferred properties and characteristics.

The use of such wetting agents as (1)—(8) and (14)—(17) with or without use of a polar or non-polar solvent produces a paste pigment which remains permanently useful and plastic. The use of such wetting agents as (9), (10), (12), (13) and (18), produces a pigment composition which partially hardens and is non-hydrolyzing under water.

An illustrative example of the compositions of such pigment compositions is as follows:—

#### EXAMPLE XV.

14—30 parts, 54% chlorine content chlorinated biphenyl; 100 parts, organophilic dodecyl ammonium bentonite pre-wetted in a bath containing the chlorinated biphenyl having a chlorine content of 54%. The pre-wetted organophilic dodecyl ammonium bentonite clay is charged into a mechanical mixer to which additional organophilic dodecyl ammonium bentonite and pigment matter may be added if desired in the proportions to produce when mixed a paste pigment having desired and predetermined consistency and color.

The amount of pre-wetting of the pigment in the above example may be such that the proportion of total wetting agent in the final paste pigment produced may, at the lowest limit, be about 15 per cent. of the weight of the paste pigment.

If the amount of pigment other than the organophilic bentonite is increased, and the amount of organophilic bentonite is decreased, a useful mass is nevertheless produced, which may be used as a constituent of inks, paints and other compositions involving organic vehicles.

All of the pigment masses of this invention are compatible with asphalt, benzyl cellulose, carnauba wax, cellulose acetate, butyrate, chlorinated rubber, the substance sold under the registered trade mark "Pliolite", coumarone and indene resins, dammar resin, ester gum, ethyl cellulose, nitro-cellulose, paraffins, chlorinated paraffins, polystyrene resins, polyisobutylene, rosin, rubber, styrene-butadiene co-polymers, vinyl resins, linseed oil, aromatics, turpentine, gasoline, benzene, diacetone, mineral spirits, xylene, esters such as amyl acetate, dibutyl phthalate, ethyl lactate and tricresyl phosphate and numerous other organic materials. They are thus adapted

to use in a wide variety of fields not open to the oil, oil-turpentine or mineral spirits paste pigments known heretofore.

Pigment masses of this invention used in paints, not only give a higher gloss and smoother coverage, with no piling or lumping, but give better coverage than the paste pigments known heretofore. For example, a paint containing 25 ounces of the 40% aluminium paste pigment of Example I per (United States of America) gallon of paint gives better coverage than a paint containing 32 ounces per gallon of a paste pigment consisting of 74% aluminium and 27% mineral spirits, all of the remaining constituents being the same.

The pigment compositions of this invention exhibit a high degree of adhesion, have high dielectric strength and low power factor, improved color and container stability and increased utility.

Numerous variations within the scope of the appended claims will be apparent to those skilled in the art in the light of the foregoing description.

Thus it can be seen that pigment compositions are provided which not only accomplish the objects set forth, but are economical, versatile, efficient and convenient to use.

What I claim is:—

1. A paste pigment comprising pigment that has been wetted and coated with a wetting agent consisting of, or containing at least 5 per cent. (by weight) of, chlorinated polyphenyl in liquid form or in solution, the proportion of the said wetting agent used in the wetting of the pigment being within the range 15—90 per cent. (by weight) of the paste pigment formed therewith.

2. A paste pigment having a soft, plastic consistency and containing ferrous or non-ferrous metal particles wetted and coalesced in a wetting agent consisting of, or containing at least 5 per cent. (by weight) of, chlorinated polyphenyl in liquid form or in solution, said chlorinated polyphenyl having a chlorine content of approximately 20—65 per cent., the wetting agent being mixed with the particles in a proportion which is within the range 15—90 per cent. (by weight) of the paste pigment.

3. A paste pigment having a soft plastic consistency and containing non-metal pigment matter wetted and coalesced in a wetting agent consisting of, or containing at least 5 per cent. (by weight) of, chlorinated polyphenyl in liquid form or in solution, said chlorinated polyphenyl having a chlorine content of approximately 20—65 per cent., the wetting agent being mixed with the

pigment matter in a proportion which is within the range 15—90 per cent. (by weight) of the paste pigment.

4. A paste pigment accoring to any  
5 one of the preceding claims, which is non-inflammable, non-oxidizing, and non-hydrolyzing and comprises pigment and liquid chlorinated biphenyl having a chlorine content of approximately 54—  
10 65 per cent. mixed in suitable proportions to form a soft or plastic pigment composition having a high solids and low volatile content.

5. A paste pigment according to Claim  
15 1 or 3, in which the wetted pigment is taken from the group consisting of natural or synthetic graphite, or carbon black.

6. A paste pigment according to Claim  
20 1 or 3, in which the wetted pigment is an organophilic cation-modified onium ben-tonite, of the character described in

British Patent No. 664,830 with or without additional metal or non-metal pigment matter.

7. A new and improved paste pigment according to Claim 1, containing pigment matter which has been pre-wetted in a cold or hot bath medium comprising liquid chlorinated polyphenyl, the pre-  
30 wetted pigment having been coalesced to form a soft or plastic paste pigment.

8. A paste pigment according to Claim 1, in which the pigment has been wetted and mixed in a wetting agent comprising  
35 chlorinated polyphenyl in liquid form with a suitable solvent, thinner, diluent or plasticizer in sufficient proportions to form a non-discoloring, paste pigment which is acid, alkali, water and corrosion  
40 resistant.

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